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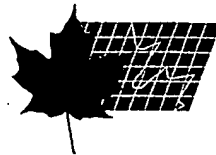
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(19) (CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) "Substituted Triazolinones and Their Use as Herbicides"

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Notice: This application is as filed and may therefore contain an
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WELTORGANISATION FÜR GEISTIGES EIGENTUM
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 INTERNATIONALE ANMELDUNG VERÖFFENTLICHT NACH DEM VERTRAG ÜBER DIE
 INTERNATIONALE ZUSAMMENARBEIT AUF DEM GEBIET DES PATENTWESENS (PCT)

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(21) Internationales Aktenzeichen: PCT/EP95/00466 (22) Internationales Anmeldedatum: 9. Februar 1995 (09.02.95) (30) Prioritätsdaten: P 44 05 614.1 22. Februar 1994 (22.02.94) DE (71) Anmelder (für alle Bestimmungsstaaten ausser US): BAYER AKTIENGESELLSCHAFT [DE/DE]; D-51368 Leverkusen (DE). (72) Erfinder; und (75) Erfinder/Anmelder (nur für US): LINKER, Karl-Heinz [DE/DE]; Albert-Schweitzer-Strasse 3, D-51377 Lev- erkusen (DE). FINDEISEN, Kurt [DE/DE]; Dünfelder Strasse 28, D-51375 Leverkusen (DE). HAAS, Wil- helm [DE/DE]; Schürgespfad 19, D-50259 Pulheim (DE). SCHALLNER, Otto [DE/DE]; Noldeweg 22, D- 40789 Monheim (DE). WROBLOWSKY, Heinz-Jürgen [DE/DE]; Virneburgstrasse 73, D-40764 Langenfeld (DE). DOLLINGER, Markus [DE/DE]; Burscheider Strasse 154b, D-51381 Leverkusen (DE). SANTEL, Hans-Joachim [DE/DE]; Grünstrasse 9a, D-51371 Leverkusen (DE).	(74) Gemeinsamer Vertreter: BAYER AKTIENGE- SELLSCHAFT; D-51368 Leverkusen (DE). (81) Bestimmungsstaaten: AU, BB, BG, BR, BY, CA, CN, CZ, FI, HU, JP, KR, KZ, LK, NO, NZ, PL, RO, RU, SK, UA, US, europäisches Patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI Patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG). Veröffentlicht Mit internationalem Recherchenbericht. Vor Ablauf der für Änderungen der Ansprüche zugelassenen Frist. Veröffentlichung wird wiederholt falls Änderungen eintreffen.	

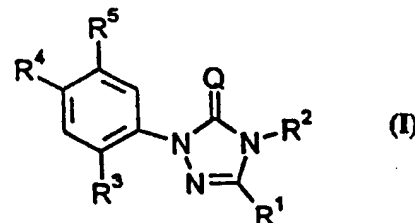
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(54) Title: SUBSTITUTED TRIAZOLINONES AND THEIR USE AS HERBICIDES

(54) Bezeichnung: SUBSTITUIERTE TRIAZOLINONE UND IHRE VERWENDUNG ALS HERBIZIDE

(57) Abstract

New substituted triazolinones have the general formula (I), in which Q stands for oxygen or sulphur, R¹ stands for halogen alkyl, R³ stands for hydrogen or halogen, R⁴ stands for cyano or nitro, R² stands for hydrogen or various substituents and R⁵ stands for various substituents. Also disclosed are methods for producing these triazolinones and their use as herbicides.



(57) Zusammenfassung

Die Erfindung betrifft neue substituierte Triazolinone der allgemeinen Formel (I), in welcher Q für Sauerstoff oder Schwefel, R¹ für Halogenalkyl, R³ für Wasserstoff oder Halogen, R⁴ für Cyano oder Nitro, R² für Wasserstoff oder verschiedene Substituenten und R⁵ für verschiedene Substituenten stehen, Verfahren zu ihrer Herstellung und ihre Verwendung als Herbizide.

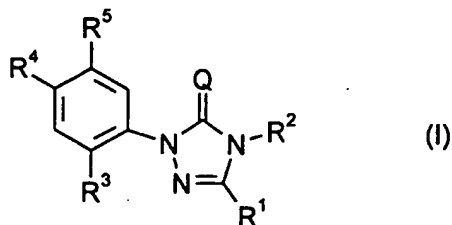
SUBSTITUTED TRIAZOLINONES AND THEIR USE AS HERBICIDES

The invention relates to new substituted triazolinones, to processes for their preparation, and to their use as herbicides.

It has been disclosed that certain substituted triazolinones, such as, for example, the compound 2-(4-cyano-2,5-difluoro-phenyl)-5-methyl-4-propargyl-2,4-dihydro-3H-1,2,4-triazol-3-one, have herbicidal properties (cf. EP-A 370332).

However, the activity of these known compounds is not satisfactory in all respects.

There have now been found the new substituted triazolinones of the general formula (I)



10 in which

Q represents oxygen or sulphur,

R¹ represents halogenoalkyl,

R² represents hydrogen, amino, cyano, alkyl, alkenyl, alkynyl, halogenoalkyl, halogenoalkenyl, halogenoalkynyl, alkoxyalkyl, alkylideneimino or in each case optionally substituted cycloalkyl or cycloalkylalkyl,

R³ represents hydrogen or halogen,

- R⁴ represents cyano or nitro, and
- R⁵ represents isocyano, thiocyanato, sulpho, halogenosulphonyl, alkylaminooxy, dialkylaminooxy, alkylideneaminooxy, cycloalkylideneaminooxy, or in each case optionally substituted cycloalkenyloxy or heterocyclyloxy, or represents one of the following groups which are bonded via nitrogen or oxygen
- 5 -NR⁶R⁷, -N=CR⁸R⁹, -O-CO-R¹⁰, -O-CS-R¹⁰, -O-CHR¹¹-P(O)(OR¹²)₂, where
- R⁶ represents hydrogen, or represents alkyl, alkenyl or alkynyl, each of which is optionally substituted by halogen, or represents the group -CO-R¹³ in which
- 10 R¹³ represents hydrogen, or represents alkyl or alkoxy, each of which is optionally substituted by halogen or alkoxy, or represents amino, alkylamino or dialkylamino, or represents in each case optionally substituted cycloalkyl, cycloalkylalkyl, aryl, arylalkyl or heterocyclyl-alkyl,
- 15 R⁷ represents the group -(CO)_n-R¹³ in which
- R¹³ has the abovementioned meaning and
- n represents the numbers 1 or 2,
- R⁸ represents hydrogen, alkyl or alkoxy,
- R⁹ represents alkoxy, alkylamino or dialkylamino,
- 20 R¹⁰ represents alkyl which is optionally substituted by halogen, or represents

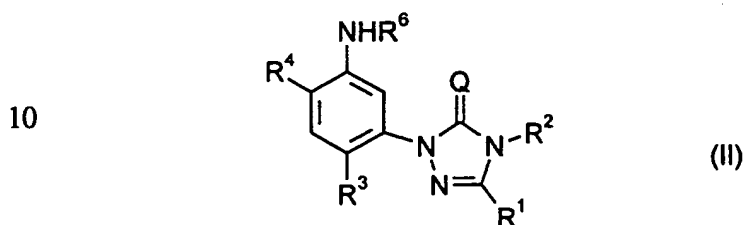
alkoxy, or represents alkylamino, or represents dialkylamino or represents optionally substituted aryl,

5 R^{11} represents alkyl or cycloalkyl, each of which is optionally substituted by halogen, or represents in each case optionally substituted aryl or heterocyclyl, and

R^{12} represents alkyl.

Furthermore, it has been found that the new substituted triazolinones of the general formula (I) are obtained when

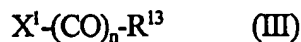
(a) aminoaryltriazolinones of the general formula (II)



in which

Q, R^1 , R^2 , R^3 , R^4 and R^6 have the abovementioned meanings

are reacted with acid halides of the general formula (III)

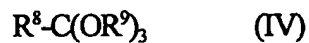


15 in which

n and R^{13} have the abovementioned meaning and

X^1 represents halogen,

or with ortho esters of the general formula (IV)



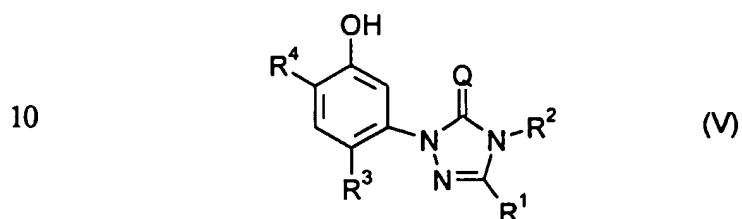
in which

5 R^8 and R^9 have the abovementioned meanings,

if appropriate in the presence of a reaction auxiliary and if appropriate in the presence of a diluent,

or when

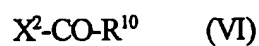
(b) hydroxyaryltriazolinones of the general formula (V)



in which

Q , R^1 , R^2 , R^3 and R^4 have the abovementioned meanings

are reacted with acid halides of the general formula (VI)



15 in which

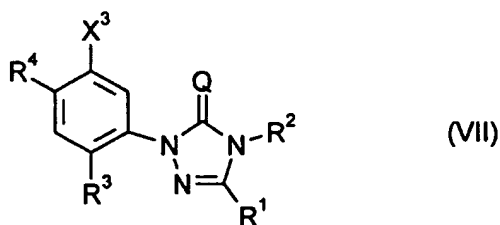
R¹⁰ has the abovementioned meaning and

X² represents halogen,

if appropriate in the presence of an acid acceptor and if appropriate in the presence of a diluent,

5 or when

(c) halogenoaryltriazolinones of the general formula (VII)



in which

Q, R¹, R², R³ and R⁴ have the abovementioned meanings and

10 X³ represents halogen,

are reacted with hydroxyalkylphosphonic esters of the general formula (VIII)



in which

R¹¹ and R¹² have the abovementioned meanings,

15 if appropriate in the presence of an acid acceptor and if appropriate in the presence of

a diluent.

Finally, it has been found that the new substituted triazolinones of the general formula (I) have interesting herbicidal properties.

Surprisingly, the substituted triazolinones of the general formula (I) according to the invention show a considerably better herbicidal activity against problem weeds in comparison with the substituted triazolinones known from the prior art, such as, for example, the compound 2-(4-cyano-2,5-difluoro-phenyl)-5-methyl-4-propargyl-2,4-dihydro-3H-1,2,4-triazol-3-one.

The invention preferably relates to compounds of the formula (I) in which

- 10 Q represents oxygen or sulphur,
R¹ represents straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, in particular fluorine, chlorine, bromine or iodine,
- 15 R² represents hydrogen, amino, cyano, straight-chain or branched alkyl having 1 to 8 carbon atoms, in each case straight-chain or branched alkenyl or alkynyl, each of which has 2 to 6 carbon atoms, straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, in particular fluorine, chlorine, bromine or iodine, or represents in each case straight-chain or branched halogenoalkenyl or halogenoalkynyl, each of which
- 20 has 2 to 6 carbon atoms and 1 to 11 identical or different halogen atoms, in particular fluorine, chlorine, bromine or iodine, or represents straight-chain or branched alkoxyalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, or represents straight-chain or branched alkylideneimino having 1 to 8 carbon atoms, or represents cycloalkyl or cycloalkylalkyl, each of which
- 25 has 3 to 8 carbon atoms in the cycloalkyl moiety and, if appropriate, 1 to

4 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted in the cycloalkyl moiety by identical or different halogen substituents, in particular fluorine, chlorine, bromine and/or iodine,

5 R^3 represents hydrogen, fluorine, chlorine, bromine or iodine,

R^4 represents cyano or nitro, and

10 R^5 represents isocyano, thiocyanato, sulpho, halogenosulphonyl, C_1 - C_6 -alkylaminooxy, di(C_1 - C_4 -alkyl)-aminooxy, C_1 - C_6 -alkylideneaminooxy or C_5 - C_6 -cycloalkylideneaminooxy, or represents C_5 - C_6 -cycloalkenyloxy, perhydrofuranyloxy or perhydropyranyloxy, each of which is optionally substituted by halogen, C_1 - C_4 -alkyl or C_1 - C_4 -alkoxy, or represents one of the following groups which are bonded via nitrogen or oxygen

$-NR^6R^7$, $-N=CR^8R^9$, $-O-CO-R^{10}$, $-O-CS-R^{10}$, $-O-CHR^{11}-P(O)(OR^{12})_2$, where

15 R^6 represents hydrogen, or represents alkyl, alkenyl or alkynyl, each of which has up to 6 carbon atoms and each of which is optionally substituted by fluorine and/or chlorine, or represents the group $-CO-R^{13}$ in which

20 R^{13} represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 6 carbon atoms and each of which is optionally substituted by fluorine, chlorine or C_1 - C_4 -alkoxy, or represents amino, or represents alkylamino or dialkylamino, each of which has 1 to 6 carbon atoms in the alkyl moieties, or represents in each case optionally substituted C_3 - C_6 -cycloalkyl, C_3 - C_6 -cycloalkyl- C_1 - C_4 -alkyl, phenyl, naphthyl, phenyl-

C₁-C₄-alkyl, furyl, thienyl or pyridyl, suitable substituents in each case being:

5 halogen, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 4 carbon atoms, in each case straight-chain or
10 branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in each case straight-chain or branched alkoxy carbonyl or
15 alkoxyiminoalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, and phenyl which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen and/or straight-chain or branched alkyl or alkoxy, each of which has 1 to 4 carbon atoms, and/or straight-chain or branched halogenoalkyl or halogenoalkoxy, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms;

R⁷ represents the group -(CO)_n-R¹³ in which

R¹³ has the meaning given above as being preferred and

20 n represents the numbers 1 or 2,

R⁸ represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 6 carbon atoms,

R⁹ represents alkoxy, alkylamino or dialkylamino, each of which has 1 to 6 carbon atoms in the alkyl groups,

5 R¹⁰ represents alkyl having 1 to 6 carbon atoms which is optionally substituted by fluorine and/or chlorine, or represents alkoxy or alkylamino or dialkylamino, each of which has 1 to 6 carbon atoms in the alkyl groups, or represents optionally substituted phenyl, preferred phenyl substituents being, again, those which have been mentioned above for R⁶ as being preferred,

10 R¹¹ represents alkyl having 1 to 6 carbon atoms or cycloalkyl having 3 to 6 carbon atoms, in each case optionally substituted by fluorine and/or chlorine, or represents in each case optionally substituted phenyl, naphthyl, furyl, thienyl or pyridyl, preferred phenyl substituents being, again, those which have been mentioned above for R⁶ as being preferred, and

 R¹² represents alkyl having 1 to 6 carbon atoms.

In particular, the invention relates to compounds of the formula (I) in which

15 Q represents oxygen or sulphur,

 R¹ represents straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in particular fluorine, chlorine, bromine or iodine,

20 R² represents hydrogen, amino, cyano, straight-chain or branched alkyl having 1 to 4 carbon atoms, in each case straight-chain or branched alkenyl or alkynyl, each of which has 3 to 4 carbon atoms, straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in particular fluorine or chlorine, in each case straight-chain or branched halogenoalkenyl or halogenoalkynyl, each of which has 3 to 4 carbon atoms and

1 to 5 identical or different halogen atoms, in particular fluorine or chlorine, or represents straight-chain or branched alkoxyalkyl having in each case 1 to 3 carbon atoms in the individual alkyl moieties, or represents straight-chain or branched alkylideneimino having 1 to 4 carbon atoms, or represents cycloalkyl or cycloalkylalkyl, each of which has 3 to 6 carbon atoms in the cycloalkyl moiety and, if appropriate, 1 to 3 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted in the cycloalkyl moiety by identical or different halogen substituents, in particular fluorine or chlorine,

10 R^3 represents hydrogen, fluorine or chlorine,

R^4 represents cyano or nitro, and

R^5 represents isocyano, thiocyanato, sulpho, chlorosulphonyl, C_1 - C_4 -alkyl-aminooxy, di- $(C_1$ - C_3 -alkyl)-aminooxy, C_1 - C_4 -alkylidene-aminooxy, C_5 - C_6 -cycloalkylideneaminooxy, C_5 - C_6 -cycloalkenyloxy, tetrahydrofuranyloxy, perhydropyranyloxy or one of the following groups which are bonded via nitrogen or oxygen

$-NR^6R^7$, $-N=CR^8R^9$, $-O-CO-R^{10}$, $-O-CS-R^{10}$, $-O-CHR^{11}-P(O)(OR^{12})_2$ where

20 R^6 represents hydrogen, or represents alkyl having 1 to 4 carbon atoms which is optionally substituted by fluorine, or represents the group $-CO-R^{13}$ in which

R^{13} represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 4 carbon atoms and each of which is optionally substituted by fluorine, chlorine, methoxy or ethoxy, or represents amino, or represents alkylamino or dialkylamino, each

of which has 1 to 4 carbon atoms in the alkyl moieties, or
 represents in each case optionally substituted cyclopropyl,
 cyclopentyl cyclohexyl, cyclopropylmethyl, cyclopentylmethyl,
 cyclohexylmethyl, phenyl, benzyl, phenylethyl, furyl, thienyl or
 5 pyridyl, suitable substituents in each case being:
 fluorine, chlorine, bromine, cyano, nitro, in each case straight-
 chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or
 alkylsulphonyl, each of which has 1 to 4 carbon atoms, in each
 case straight-chain or branched halogenoalkyl, halogenoalkoxy,
 10 halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkyl-
 sulphonyl, each of which has 1 to 4 carbon atoms and 1 to
 9 identical or different halogen atoms, in particular fluorine
 and/or chlorine atoms, in each case straight-chain or branched
 alkoxycarbonyl or alkoxyiminoalkyl, each of which has 1 to
 15 4 carbon atoms in the individual alkyl moieties, and phenyl
 which is optionally monosubstituted or polysubstituted by
 identical or different substituents from the series consisting of
 fluorine, chlorine, bromine and/or straight-chain or branched
 alkyl or alkoxy, each of which has 1 to 4 carbon atoms, and/or
 20 straight-chain or branched halogenoalkyl or halogenoalkoxy, each
 of which has 1 to 4 carbon atoms and 1 to 9 identical or
 different halogen atoms, in particular fluorine and/or chlorine
 atoms;

R^7 represents the group $-(CO)_n-R^{13}$ in which

25 R^{13} has the meaning given above as being particularly preferred and

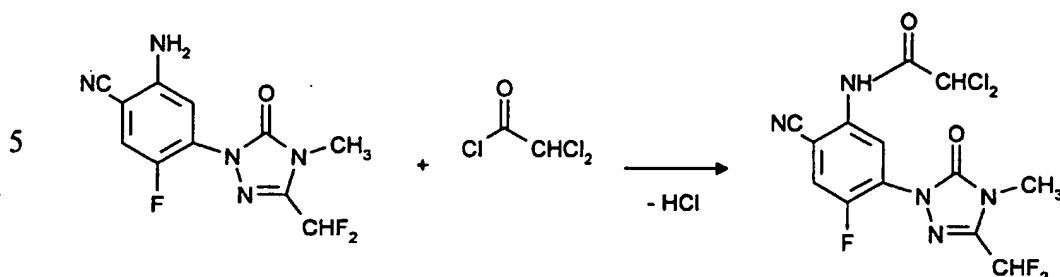
n represents the numbers 1 or 2,

- R^8 represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 4 carbon atoms,
- R^9 represents alkoxy, alkylamino or dialkylamino, each of which has 1 to 4 carbon atoms in the alkyl groups,
- 5 R^{10} represents alkyl having 1 to 4 carbon atoms which is optionally substituted by fluorine and/or chlorine, or represents alkoxy or alkylamino or dialkylamino, each of which has 1 to 4 carbon atoms in the alkyl groups, or represents optionally substituted phenyl, preferred phenyl substituents being, again, those which have been mentioned
10 above for R^6 as being particularly preferred,
- R^{11} represents alkyl having 1 to 4 carbon atoms or cycloalkyl having 3 to 6 carbon atoms, in each case optionally substituted by fluorine and/or chlorine, or represents in each case optionally substituted phenyl, naphthyl, furyl, thienyl or pyridyl, preferred phenyl substituents being,
15 again, those which have been mentioned above for R^6 as being particularly preferred, and
- R^{12} represents alkyl having 1 to 4 carbon atoms.

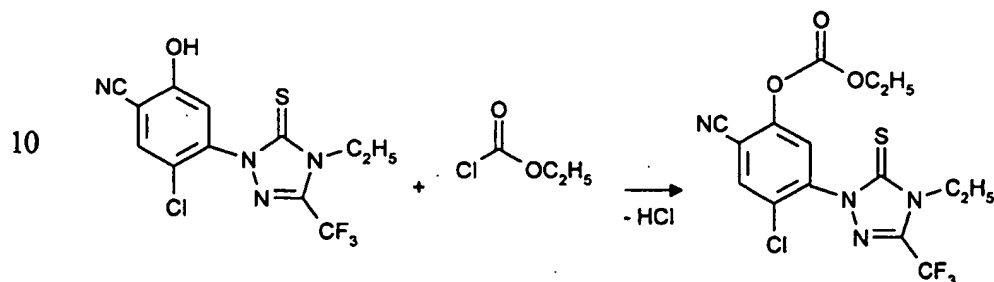
The abovementioned definitions of radicals, in general or where preferred ranges are given, apply to the end products of the formula (I) and, analogously to the starting
20 materials or intermediates required in each case for the preparation.

These definitions of radicals can be combined with each other as desired, that is to say combinations between the abovementioned ranges of preferred compounds are also possible.

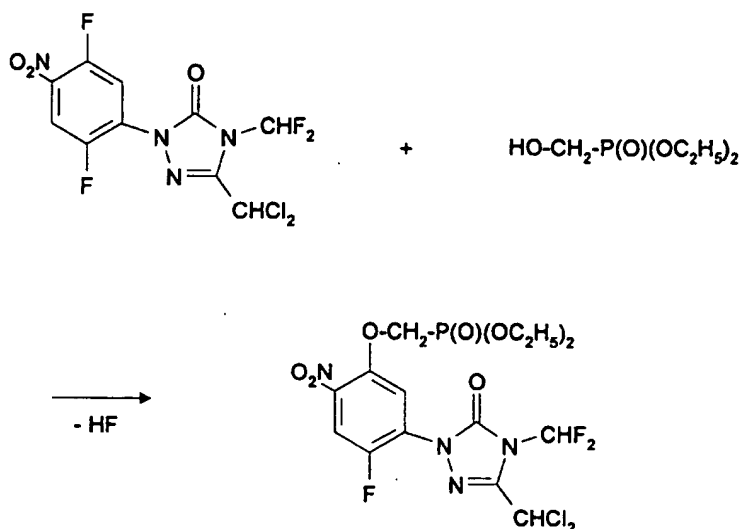
If, for example, 2-(5-amino-4-cyano-2-fluoro-phenyl)-5-difluoromethyl-4-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one and dichloroacetyl chloride are used as starting substances for carrying out process (a) according to the invention, the course of the reaction can be outlined by the following equation



If, for example, 2-(2-chloro-4-cyano-5-hydroxy-phenyl)-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazole-3-thione and ethyl chloroformate are used as starting substances for carrying out process (b) according to the invention, the course of the reaction can be outlined by the following equation



If, for example, 2-(2,5-difluoro-4-nitro-phenyl)-5-dichloromethyl-4-difluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one and diethyl hydroxymethane-phosphonate are used as starting substances for carrying out process (c) according to the invention, the course of the reaction can be outlined by the following equation:



Formula (II) provides a general definition of the aminoaryltriazolinones to be used as starting substances in process (a) according to the invention for the preparation of the compounds of the general formula (I). In formula (II), Q, R¹, R², R³, R⁴ and R⁶ preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds of the formula (I) as being preferred, or particularly preferred, for Q, R¹, R², R³, R⁴ and R⁶.

The starting substances of the formula (II) were hitherto not known from the literature; however, they are the subject-matter of earlier, non-prior-published patent applications.

- 10 The aminoaryltriazolinones of the general formula (II) are obtained when halogenoaryltriazolinones of the general formula (VIII) - above - are reacted with ammonia at temperatures between 0°C and 150°C, if appropriate in the presence of a diluent, such as, for example, dimethyl sulphoxide (cf. the preparation examples).

15 Formula (III) provides a general definition of the acid halides furthermore optionally to be used as starting substances in process (a) according to the invention. In formula (III), n and R¹³ preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds of

the formula (I) as being preferred, or particularly preferred, for n and R¹³.

X¹ preferably represents fluorine, chlorine or bromine, in particular chlorine.

The starting substances of the formula (III) are known chemicals for organic synthesis.

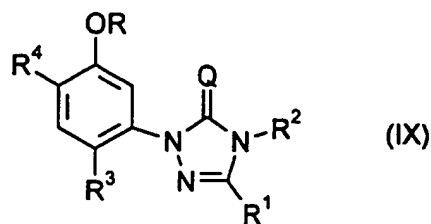
5 Formula (IV) provides a general definition of the ortho esters furthermore optionally to be used as starting substances in process (a) according to the invention. In formula (IV), R⁸ and R⁹ preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds of the formula (I) as being preferred, or particularly preferred, for R⁸ and R⁹.

The starting substances of the formula (IV) are known chemicals for organic synthesis.

10 Formula (V) provides a general definition of the hydroxyaryltriazolinones to be used as starting substances in process (b) according to the invention for the preparation of the compounds of the general formula (I). In formula (V), Q, R¹, R², R³ and R⁴ preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds of the formula (I) as being
15 preferred, or particularly preferred, for Q, R¹, R², R³ and R⁴.

The starting substances of the formula (V) were hitherto not known from the literature; however, they are subject-matter of an earlier, non-prior-published patent application (cf. DE-P 4238125/LeA 29445 dated 12.11.1992).

20 The hydroxyaryltriazolinones of the general formula (V) are obtained when alkoxyaryltriazolinones of the general formula (IX)



in which

Q, R¹, R², R³ and R⁴ have the abovementioned meanings and

R represents alkyl (preferably methyl or ethyl)

- 5 are reacted with with a dealkylating agent, such as, for example, boron(III) bromide, at temperatures of between 0°C and 50°C in the presence of a diluent, such as, for example, methylene chloride, and the product is worked up in the customary manner (cf. the preparation examples).

- 10 Formula (VI) provides a general definition of the acid halides furthermore to be used as starting substances in process (b) according to the invention. In formula (VI), R¹⁰ preferably, or in particular, has the meaning which has already been mentioned above in connection with the description of the compounds of the formula (I) as being preferred, or particularly preferred, for R¹⁰;

X² preferably represents fluorine, chlorine or bromine, in particular chlorine.

- 15 The starting substances of the formula (VI) are known chemicals for organic synthesis.

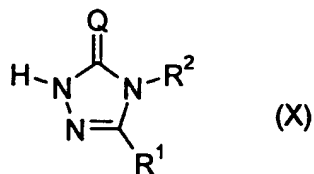
Formula (VII) provides a general definition of the halogenoaryltriazolinones to be used as starting substances in process (c) according to the invention for the preparation of the compounds of the general formula (I). In formula (VII), Q, R¹, R², R³ and R⁴ preferably, or in particular, have those meanings which have already been mentioned

above in connection with the description of the compounds of the formula (I) as being preferred, or particularly preferred, for Q, R¹, R², R³ and R⁴.

X³ preferably represents fluorine or chlorine, in particular fluorine.

5 The starting substances of the formula (VII) were hitherto not known from the literature; however, they are the subject-matter of an earlier, non-prior-published patent application.

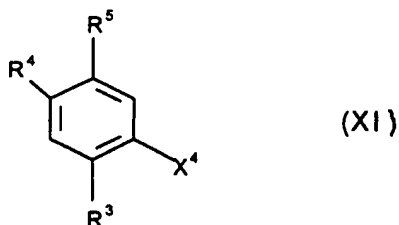
The halogenoaryltriazolinones of the general formula (VII) are obtained when triazolinones of the general formula (X)



10 in which

Q, R¹ and R² have the abovementioned meanings

are reacted with halogenoarenes of the general formula (XI)



in which

15 R³, R⁴ and R⁵ have the abovementioned meanings and

X⁴ represents halogen, preferably fluorine or chlorine, in particular fluorine,

at temperatures between 0°C and 150°C in the presence of a diluent, such as, for example, dimethyl sulphoxide and in the presence of an acid acceptor, such as, for example, potassium carbonate, and the products are worked up in the customary
5 manner (cf. the preparation examples).

Formula (VIII) provides a general definition of the hydroxyalkylphosphonic esters furthermore to be used as starting substances in process (c) according to the invention. In formula (VIII), R¹¹ and R¹² preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the
10 compounds of the formula (I) as being preferred, or particularly preferred, for R¹¹ and R¹².

The starting substances of the formula (VIII) are known chemicals for organic synthesis.

Suitable diluents for carrying out processes (a), (b) and (c) according to the invention
15 are the customary organic solvents. These include, in particular, aliphatic, alicyclic or aromatic, optionally halogenated hydrocarbons, such as, for example, benzene, toluene, xylene, chlorobenzene, dichlorobenzene, petroleum ether, hexane, cyclohexane, dichloromethane, chloroform, tetrachloromethane; ethers, such as diethyl ether, diisopropyl ether, dioxane, tetrahydrofuran, ethylene glycol dimethyl ether or ethylene
20 glycol diethyl ether, ketones, such as acetone, butanone or methyl isobutyl ketone; nitriles, such as acetonitrile, propionitrile or benzonitrile; amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; esters, such as methyl acetate or ethyl acetate, sulphoxides, such as dimethyl sulphoxide, alcohols, such as methanol,
25 ethanol, n- or i-propanol, ethylene glycol monomethyl ether, ethylene glycol monoethyl ether, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, their

mixtures with water, or pure water.

- Process (a) according to the invention is preferably carried out in the presence of a suitable reaction auxiliary. Reaction auxiliaries which are suitable for the reaction with the acid halides of the formula (III) are all customary inorganic or organic bases. These include, for example, the hydrides, hydroxides, amides, alcoholates, acetates, carbonates or hydrogen carbonates of alkaline earth metals or alkali metals, such as, for example, sodium hydride, sodium amide, sodium methylate, sodium ethylate, potassium tert-butylate, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium hydrogen carbonate, sodium hydrogen carbonate or ammonium carbonate, and also basic organic nitrogen compounds such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, pyridine, N-methylpiperidine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).
- Reaction auxiliaries which are generally employed for the reaction with the ortho esters of the formula (IV) are acidic catalysts. Suitable catalysts are, preferably, strong protonic acids, such as, for example, hydrochloric acid or hydrogen chloride, sulphuric acid, methanesulphonic acid, benzenesulphonic acid and p-toluenesulphonic acid.

- When carrying out process (a) according to the invention, the reaction temperatures can be varied within a substantial range. In general, the process is carried out at temperatures between 0°C and 150°C, preferably at temperatures between 10°C and 100°C.

- Process (a) according to the invention is generally carried out under atmospheric pressure. However, it is also possible to carry out the process under elevated or reduced pressure, in general between 0.1 bar and 10 bar.

To carry out process (a) according to the invention, the starting substances required in each case are generally employed in approximately equimolar amounts. However, it is also possible to use a larger excess of one of the two components employed in each case. In general, the reactions are carried out in a suitable diluent in the presence of a reaction auxiliary, and the reaction mixture is stirred for several hours at the temperature required in each case. Working-up in the process according to the invention is carried out in each case by customary methods (cf. the preparation examples).

Processes (b) and (c) according to the invention are preferably carried out in the presence of a suitable acid acceptor. Suitable acid acceptors are all customary inorganic or organic bases. These include, for example, the hydrides, hydroxides, amides, alcoholates, acetates, carbonates or hydrogen carbonates of alkaline earth metals or alkali metals, such as, for example, sodium hydride, sodium amide, sodium methylate, sodium ethylate, potassium tert-butyrate, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium hydrogen carbonate, sodium hydrogen carbonate or ammonium carbonate, and also basic organic nitrogen compounds such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, pyridine, N-methylpiperidine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

When carrying out processes (b) and (c) according to the invention, the reaction temperatures can be varied within a substantial range. In general, the processes are carried out at temperatures between 0°C and 100°C, preferably at temperatures between 10°C and 80°C.

Processes (b) and (c) according to the invention are generally carried out under normal pressure. However, it is also possible to carry out the processes under elevated or reduced pressure, in general between 0.1 bar and 10 bar.

To carry out processes (b) and (c) according to the invention, the starting substances required in each case are generally employed in approximately equimolar amounts. However, it is also possible to use one of the two components employed in each case in a larger excess. The reactions are generally carried out in a suitable diluent in the presence of an acid acceptor, and the reaction mixture is stirred for several hours at the temperature required in each case. Working-up in the process according to the invention is carried out in each case by customary methods (cf. the preparation examples).

The active compounds according to the invention can be used as defoliants, desiccants, agents for destroying broad-leaved plants and, especially, as weed-killers. By weeds, in the broadest sense, there are to be understood all plants which grow in locations where they are undesired. Whether the substances according to the invention act as total or selective herbicides depends essentially on the amount used.

The active compounds according to the invention can be used, for example, in connection with the following plants:

Dicotyledon weeds of the genera: Sinapis, Lepidium, Galium, Stellaria, Matricaria, Anthemis, Galinsoga, Chenopodium, Urtica, Senecio, Amaranthus, Portulaca, Xanthium, Convolvulus, Ipomoea, Polygonum, Sesbania, Ambrosia, Cirsium, Carduus, Sonchus, Solanum, Rorippa, Rotala, Lindernia, Lamium, Veronica, Abutilon, Emex, Datura, Viola, Galeopsis, Papaver, Centaurea, Trifolium, Ranunculus and Taraxacum.

Dicotyledon cultures of the genera: Gossypium, Glycine, Beta, Daucus, Phaseolus, Pisum, Solanum, Linum, Ipomoea, Vicia, Nicotiana, Lycopersicon, Arachis, Brassica, Lactuca, Cucumis and Cucurbita.

Monocotyledon weeds of the genera: Echinochloa, Setaria, Panicum, Digitaria, Phleum, Poa, Festuca, Eleusine, Brachiaria, Lolium, Bromus, Avena, Cyperus,

Sorghum, Agropyron, Cynodon, Monochoria, Fimbristylis, Sagittaria, Eleocharis, Scirpus, Paspalum, Ischaemum, Sphenoclea, Dactyloctenium, Agrostis, Alopecurus and Apera.

- Monocotyledon cultures of the genera: Oryza, Zea, Triticum, Hordeum, Avena, Secale,
5 Sorghum, Panicum, Saccharum, Ananas, Asparagus and Allium.

However, the use of the active compounds according to the invention is in no way restricted to these genera, but also extends in the same manner to other plants.

- The compounds are suitable, depending on the concentration, for the total combating of weeds, for example on industrial terrain and rail tracks, and on paths and squares
10 with or without tree plantings. Equally, the compounds can be employed for combating weeds in perennial cultures, for example afforestations, decorative tree plantings, orchards, vineyards, citrus groves, nut orchards, banana plantations, coffee plantations, tea plantations, rubber plantations, oil palm plantations, cocoa plantations, soft fruit plantings and hopfields, in lawns, turf and pasture-land, and for the selective combating
15 of weeds in annual cultures.

The compounds (I) according to the invention are particularly suitable for selectively combating monocotyledon and dicotyledon weeds in monocotyledon and dicotyledon cultures, such as, for example, in wheat and barley, by the pre-emergence and also the post-emergence method.

- 20 To a certain extent, the compounds of the formula (I) also have a fungicidal activity, for example against pyricularia oryzae, phytophthora infestans and venturia inaequalis.

The active compounds can be converted into the customary formulations, such as solutions, emulsions, wettable powders, suspensions, powders, dusting agents, pastes, soluble powders, granules, suspension-emulsion concentrates, natural and synthetic

materials impregnated with active compound, and very fine capsules in polymeric substances.

These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents and/or solid carriers, optionally with
5 the use of surface-active agents, that is emulsifying agents and/or dispersing agents and/or foam-forming agents.

In the case of the use of water as an extender, organic solvents can, for example, also be used as auxiliary solvents. As liquid solvents, there are suitable in the main: aromatics, such as xylene, toluene or alkyl naphthalenes, chlorinated aromatics and
10 chlorinated aliphatic hydrocarbons, such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons, such as cyclohexane or paraffins, for example petroleum fractions, mineral and vegetable oils, alcohols, such as butanol or glycol as well as their ethers and esters, ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents, such as
15 dimethylformamide and dimethyl sulphoxide, as well as water.

As solid carriers there are suitable:

for example ammonium salts and ground natural minerals, such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as highly disperse silica, alumina and silicates, as solid carriers for
20 granules there are suitable: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks; as emulsifying and/or foam-forming agents there are suitable: for example non-ionic and anionic emulsifiers, such as polyoxyethylene
25 fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates as well as albumen hydrolysis

products; as dispersing agents there are suitable: for example lignin-sulphite waste liquors and methylcellulose.

Adhesives such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latexes, such as gum arabic, polyvinyl alcohol and
5 polyvinyl acetate, as well as natural phospholipids, such as cephalins and lecithins, and synthetic phospholipids, can be used in the formulations. Further additives can be mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide, Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo
10 dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations in general comprise between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

For combating weeds, the active compounds according to the invention, as such or in
15 the form of their formulations, can also be used as mixtures with known herbicides, finished formulations or tank mixes being possible.

Suitable herbicides for the mixtures are known herbicides, for example anilides such as, for example, diflufenican and propanil; arylcarboxylic acids such as, for example, dichloropicolinic acid, dicamba and picloram; aryloxyalkanoic acids such as, for
20 example, 2,4 D, 2,4 DB, 2,4 DP, fluroxypyr, MCPA, MCPP and triclopyr; aryloxyphenoxy-alkanoic esters such as, for example, diclofop-methyl, fenoxaprop-ethyl, fluazifop-butyl, haloxyfop-methyl and quizalofop-ethyl; azinones such as, for example, chloridazon and norflurazon; carbamates such as, for example, chlorpropham, desmedipham, phenmedipham and propham; chloroacetanilides such as, for example,
25 alachlor, acetochlor, butachlor, metazachlor, metolachlor, pretilachlor and propachlor;

dinitroanilines such as, for example, oryzalin, pendimethalin and trifluralin; diphenyl ethers such as, for example, acifluorfen, bifenox, fluoroglycofen, fomesafen, halosafen, lactofen and oxyfluorfen; ureas such as, for example, chlortoluron, diuron, fluometuron, isoproturon, linuron and methabenzthiazuron; hydroxylamines such as, for example, alloxydim, clethodim, cycloxydim, sethoxydim and tralkoxydim; imidazolinones such as, for example, imazethapyr, imazamethabenz, imazapyr and imazaquin; nitriles such as, for example, bromoxynil, dichlobenil and ioxynil; oxyacetamides such as, for example, mefenacet; sulphonylureas such as, for example, amidosulfuron, bensulfuron-methyl, chlorimuron-ethyl, chlorsulfuron, cinosulfuron, metsulfuron-methyl, nicosulfuron, primisulfuron, pyrazosulfuron-ethyl, thifensulfuron-methyl, triasulfuron and tribenuron-methyl; thiocarbamates such as, for example, butylate, cycloate, di-allate, EPTC, esprocarb, molinate, prosulfocarb, thiobencarb and tri-allate; triazines such as, for example, atrazine, cyanazine, simazine, simetryn, terbutryn and terbutylazine; triazinones such as, for example, hexazinone, metamiltron and metribuzin; others such as, for example, aminotriazole, benfuresate, bentazone, cinmethylin, clomazone, clopyralid, difenzoquat, dithiopyr, ethofumesate, fluorochloridone, glufosinate, glyphosate, isoxaben, pyridate, quinchlorac, quinmerac, sulposate and tridiphane.

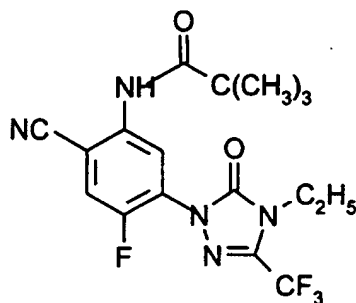
Mixtures with other known active compounds, such as fungicides, insecticides, acaricides, nematicides, bird repellants, plant nutrients and agents which improve soil structure, are also possible.

The active compounds can be used as such, in the form of their formulations or in the use forms prepared therefrom by further dilution, such as ready-to-use solutions, suspensions, emulsions, powders, pastes and granules. They are used in the customary manner, for example by watering, spraying, atomizing or scattering.

The active compounds according to the invention can be applied either before or after emergence of the plants. They can also be incorporated into the soil before sowing.

The amount of active compound used can vary within a substantial range. It depends essentially on the nature of the desired effect. In general, the amounts used are between 10 g and 10 kg of active compound per hectare of soil surface, preferably between 50 g and 5 kg per ha.

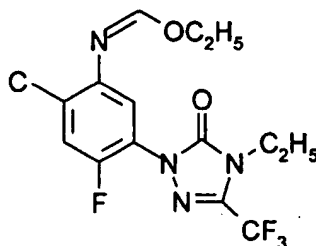
- 5 The preparation and use of the active compounds according to the invention can be seen from the following examples.

Preparation Examples:Example 1

(Process (a))

- 5 1.01 g (0.01 mol) of triethylamine together with 3.15 g (0.01 mol) of 2-(2-fluoro-4-cyano-5-amino-phenyl)-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one in 50 ml of acetonitrile are introduced into the reaction vessel, and the stirred mixture is treated with 1.21 g (0.01 mol) of trimethylacetyl chloride, stirred for 5 hours at room temperature (20°C) and concentrated. The residue is stirred with water, filtered off with
10 suction and purified over a silica gel [lacuna] (eluent: cyclohexane/ethyl acetate 3:1).

1.7 g (43 % of theory) of 2-[2-fluoro-4-cyano-5-(tert-butyl-carbonylamino)-phenyl]-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 149°C are obtained.

Example 2

15

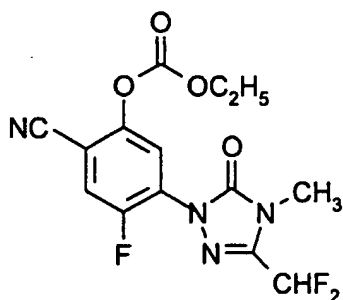
(Process (a))

3.15 g (0.01 mol) of 2-(2-fluoro-4-cyano-5-amino-phenyl)-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one and 0.01 g of p-toluenesulphonic acid in 20 ml of triethyl orthoformate are stirred for 20 minutes at reflux temperature. When cold, the
5 solution is concentrated and recrystallized from a small amount of isopropanol.

2 g (54 % of theory) of 2-(2-fluoro-4-cyano-5-ethoxymethyleneaminophenyl)-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 91°C are obtained.

Example 3

10



(Process (b))

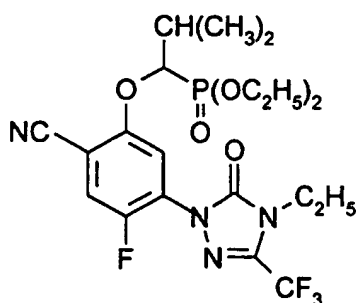
2 g (0.007 mol) of 2-(2-fluoro-4-cyano-5-hydroxy-phenyl)-4-methyl-5-difluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one in 50 ml of acetonitrile are treated with 0.23 g (0.0077 mol) of sodium hydride (80 % in paraffin), and the mixture is stirred for
15 20 minutes at 20°C; 0.84 g (0.0077 mol) of ethyl chloroformate is subsequently added, and the mixture is stirred for 8 hours at room temperature (20°C). After concentration, the mixture is stirred with water, acidified with concentrated hydrochloric acid, and extracted three times using dichloromethane, and the organic phase is dried over sodium sulphate and concentrated. For purification, the mixture is chromatographed
20 over silica gel (eluent: cyclohexane/ethyl acetate 1:1).

0.8 g (32 % of theory) of 2-[2-fluoro-4-cyano-5-(ethoxy-carbonyloxy)-phenyl]-4-methyl-5-difluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one is obtained as an oil.

¹H NMR (CDCl₃): 3.50; 4.35-4.43; 7.55-7.58 ppm

Example 4

5



(Process (c))

6.3 g (0.03 mol) of diethyl 1-hydroxy-iso-butyl-phosphonate in 100 ml of acetonitrile are treated with 0.9 g (0.03 mol) of sodium hydride (80 % in paraffin) and the mixture is stirred for 20 minutes at 20°C. After 3.15 g (0.01 mol) of 2-(2,5-difluoro-4-cyanophenyl)-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one have been added, the mixture is stirred for 12 hours at room temperature (20°C). The mixture is stirred with water and the product which has precipitated is filtered off with suction and recrystallized from cyclohexane.

3 g (59 % of theory) of 2-[2-fluoro-4-cyano-5-(1-diethoxyphosphoryl-iso-butoxy)-phenyl]-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 113°C are obtained.

Other examples of compounds of the formula (I) which can be prepared analogously to Examples 1 to 4 and following the general description of the preparation process according to the invention are those listed in Table 1 below.

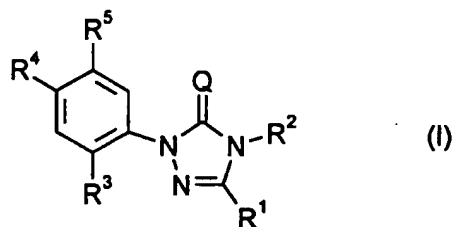
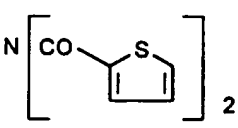
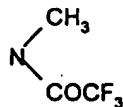


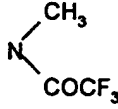
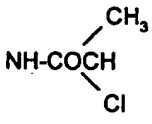
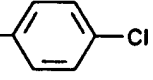
Table 1: Examples of the compounds of the formula (I)

Ex. No.	R ¹	R ²	R ³	R ⁴	R ⁵	Q	Physical properties
5	CF ₃	CH ₃	F	CN	NH-CO-C ₂ H ₅	O	m.p.: 132°C
6	CF ₃	C ₂ H ₅	F	CN	N 	O	m.p.: 107°C (decomp.)
7	CF ₃	CH ₃	F	CN		O	m.p.: 123°C
8	CF ₃	C ₂ H ₅	F	CN	NH-COCF ₃	O	m.p.: 146°C
9	CF ₃	CH ₃	F	CN	NH-COCH ₃	O	m.p.: 155°C
10	CF ₃	CH ₃	F	CN	NH-COCH(CH ₃) ₂	O	¹ H NMR (CDCl ₃ , d): = 1.14; 2.67; 3.36; 7.81; 8.15; 10.27




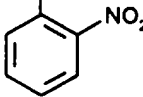
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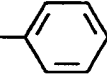
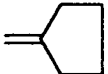
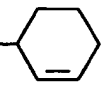
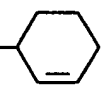
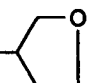
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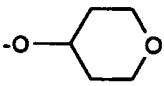
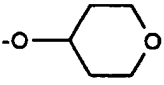
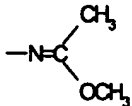
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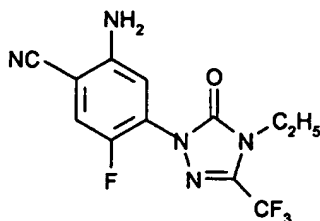
	Ex. No.	R ¹	R ²	R ³	R ⁴	R ⁵	Q	Physical properties
5	11	CF ₃	C ₂ H ₅	F	CN	NH-COCOC(CH ₃) ₃	O	m.p.: 176°C
	12	CF ₃	C ₂ H ₅	F	CN	NH-CO-NH ₂	O	m.p.: >250°C
	13	CF ₃	CH ₃	F	CN		S	¹ H NMR (CDCl ₃ , d): 3.36. 3.70
	14	CF ₃	CH ₃	F	CN	N(COCH ₃) ₂	O	¹ H NMR (CDCl ₃ , d): 2.27; 3.38; 8.06; 8.38
	15	CF ₃	C ₂ H ₅	F	CN	NH-COCH ₃	O	m.p.: 169°C
10	16	CF ₃	C ₂ H ₅	F	CN		O	¹ H NMR (CDCl ₃ , d): 1.85-1.8; 3.90- 3.98; 8.93-8.9
	17	CF ₃	C ₂ H ₅	F	CN	NH-COC(CH ₃) ₂ CH ₂ Cl	O	m.p.: 149°C
	18	CF ₃	C ₂ H ₅	F	CN	NH-COCH ₂ -Cl	O	m.p.: 121°C
	19	CF ₃	C ₂ H ₅	F	CN	N(COCH ₃) ₂	O	m.p.: 112°C (decomp.)
	20	CHF ₂	C ₂ H ₅	Cl	CN	NH-CO- 	S	
15	21	CHF ₂	CH ₃	F	CN	NH-CO-NH ₂	O	
	22	CHF ₂	CH ₃	F	NO ₂	N(COCH ₃) ₂	O	
	23	CF ₃	CH ₃	F	NO ₂	NH-COC(CH ₃) ₃	O	

	Ex. No.	R ¹	R ²	R ³	R ⁴	R ⁵	Q	Physical properties
	24	CHF ₂	CH ₃	F	CN	N=CH-OC ₂ H ₅	O	m.p.: 149°C
	25	CF ₃	C ₂ H ₅	F	CN	$\begin{array}{c} \text{CH}_3 \\ \diagup \\ \text{N}=\text{C} \\ \diagdown \\ \text{OC}_2\text{H}_5 \end{array}$	O	¹ H NMR (CDCl ₃ , d): 1.95; 3.90-3.96; 7.12-7.15
5	26	CF ₃	C ₂ H ₅	F	CN	N=C(OCH ₃) ₂	O	m.p.: 139°C
	27	CF ₃	C ₂ H ₅	F	CN	N-CH-N(CH ₃) ₂	O	
	28	CF ₃	CH ₃	Cl	CN	N=C(OCH ₃) ₂	O	
	29	CF ₃	CH ₃	F	CN	N=C(OC ₂ H ₅) ₂	S	
	30	CF ₃	C ₂ H ₅	F	NO ₂	N=CH-OC ₂ H ₅	O	
10	31	CHF ₂	CH ₃	F	CN	$\begin{array}{c} \text{CH}_3 \\ \diagup \\ \text{N}=\text{C} \\ \diagdown \\ \text{OC}_2\text{H}_5 \end{array}$	S	
	32	CF ₃	C ₂ H ₅	F	CN	-OCOOC ₂ H ₅	O	¹ H NMR (CDCl ₃ , d): 3.90-4.00; 4.35-4.45; 7.58- 7.60
	33	CF ₃	CH ₃	F	CN	-OCOOCH ₂ -CH ₂ -Cl	O	
	34	CHF ₂	CH ₃	Cl	CN	-OCOOC ₂ H ₅	O	
	35	CF ₃	CH ₃	F	NO ₂	-OCOO C ₂ H ₅	O	
15	36	CF ₃	C ₂ H ₅	F	CN	-OCOOC ₂ H ₅	S	
	37	CF ₃	C ₂ H ₅	F	CN	-OCH ₂ -PO(OC ₂ H ₅) ₂	O	m.p.: 89°C

	Ex. No.	R ¹	R ²	R ³	R ⁴	R ⁵	Q	Physical properties
	38	CF ₃	CH ₃	F	CN	-OCH ₂ -PO(OC ₂ H ₅) ₂	O	
	39	CF ₃	CH ₃	F	CN	<div>-OCH-PO(OC₂H₅)₂ CH₃</div>	O	
5	40	CHF ₂	CH ₃	F	CN	<div>-OCH-PO(OC₂H₅)₂ CH₃</div>	O	
	41	CF ₃	C ₂ H ₅	F	CN	<div>-OCH-PO(OC₂H₅)₂ CH₃</div>	O	m.p.: 90°C
	42	CF ₃	C ₂ H ₅	F	CN	<div>-OCH-PO(OC₂H₅)₂ </div>	O	
	43	CF ₃	C ₂ H ₅	F	CN	<div>-OCH-PO(OC₂H₅)₂ </div>	O	
	44	CF ₃	C ₂ H ₅	F	CN	<div>-OCH-PO(OC₂H₅)₂ </div>	O	
10	45	CF ₃	C ₂ H ₅	F	CN	<div>-OCH-PO(OC₂H₅)₂ </div>	O	
	46	CF ₃	CH ₃	F	CN	-NH-CO-C ₂ H ₅	O	
	47	CF ₃	C ₂ H ₅	F	CN	N=CH-N(CH ₃) ₂	O	m.p.: 155°C

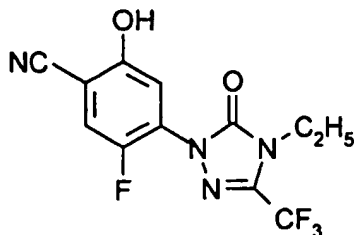
	Ex. No.	R ¹ ..	R ²	R ³	R ⁴	R ⁵	Q	Physical properties
5	48	CF ₃	C ₂ H ₅	F	CN	NH-COOC ₂ H ₅	O	m.p.: 112°C
	49	CF ₃	C ₂ H ₅	F	CN	NH-COCOOC ₂ H ₅	O	m.p.: 133°C
	50	CF ₃	C ₂ H ₅	F	CN	NH-CHO	O	m.p.: 156°C
	51	CF ₃	CH ₃	F	NO ₂	NH-COCF ₃	O	
	52	CHF ₂	CH ₃	Cl	CN	N(COCH ₃) ₂	O	
	53	CF ₃	CH ₃	F	CN	NH-CO- 	O	
10	54	CF ₃	CH ₃	F	CN	-SCN	O	n _D ²⁰ = 1.5221
	55	CF ₃	CH ₃	F	CN	-SO ₂ Cl	O	m.p.: 205°C
	56	CF ₃	CH ₃	F	CN	-O-N= 	O	m.p.: 53°C
	57	CF ₃	CH ₃	F	CN	-O-N(C ₂ H ₅) ₂	S	(amorphous)
	58	CF ₃	CH ₃	F	CN	-O-N(C ₂ H ₅) ₂	O	m.p.: 78°C
	59	CF ₃	CH ₃	F	CN	-O- 	S	¹ H NMR (CDCl ₃ , δ): 1.5-2.3; 3.70; 5.0; 5.85; 6.05; 7.75; 8.15
15	60	CF ₃	CH ₃	F	CN	-O- 	O	m.p.: 66°C
	61	CF ₃	CH ₃	F	CN	-O- 	O	m.p.: 86°C

Ex. No.	R ¹	R ²	R ³	R ⁴	R ⁵	Q	Physical properties
62	CF ₃	CH ₃	F	CN		S	¹ H NMR (CDCl ₃ , d): 1.65; 2.0; 3.5; 3.70; 3.85; 4.75; 7.75; 8.2
63	CF ₃	CH ₃	F	CN		O	mp.: 128°C
5 64	CF ₃	CH ₃	F	CN	-OCS-N(CH ₃) ₂	O	¹ H NMR (CDCl ₃ , δ): 3.40; 3.45; 3.50; 7.52-7.60 ppm;
65	CF ₃	C ₂ H ₅	F	CN		O	¹ H NMR (CDCl ₃ , δ): 1.95; 3.87; 3.90-3.98 ppm

Starting substances of the formula (II):Example (II-I):

9.54 g (0.03 mol) of 2-(2,5-difluoro-4-cyano-phenyl)-4-ethyl-5-trifluoromethyl-
 5 2,4-dihydro-3H-1,2,4-triazol-3-one in 150 ml of dimethyl sulphoxide are heated to
 120°C, and ammonia gas is passed in for 20 hours. When cold, the solution is stirred
 on ice-water, the product which has precipitated is filtered off with suction and washed
 with water, and the residue is recrystallized from isopropanol.

5.8 g (61 % of theory) of 2-(2-fluoro-4-cyano-5-aminophenyl)-4-ethyl-
 10 5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 193°C are
 obtained.

Starting substances of the formula (V):Example (V-1)

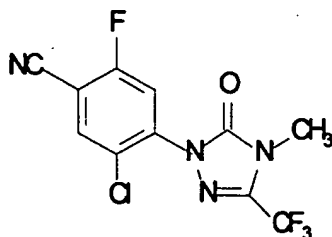
4.95 g (0.015 mol) of 2-(2-fluoro-4-cyano-5-methoxy-phenyl)-4-ethyl-5-trifluoromethyl-
 15 2,4-dihydro-3H-1,2,4-triazol-3-one in 200 ml of methylene chloride are introduced into

the reaction vessel at 10°C, and a one-molar solution of 45 g (0.045 mol) of boron(III) bromide in methylene chloride is added dropwise. The reaction mixture is stirred for 12 hours at 20°C and then treated with 100 ml of water. After the mixture has been stirred for ten minutes, the organic phase is separated off, washed with water, dried
5 using sodium sulphate and filtered. The solvent is carefully removed from the filtrate by distillation under a water pump vacuum.

3.1 g (65 % of theory) of 2-(2-fluoro-4-cyano-5-hydroxy-phenyl)-4-ethyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 195°C are obtained.

10 Starting substances of the formula (VII):

Example (VII-1)

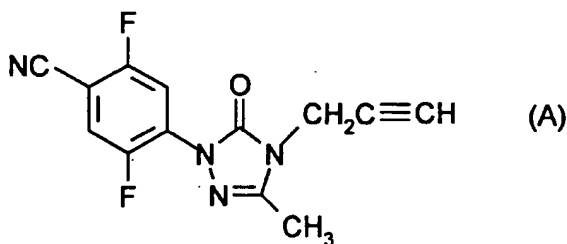


5.3 g (0.038 mol) of potassium carbonate are added at room temperature to 5.3 g (0.032 mol) of 4-methyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one (cf. for
15 example, US 3.780.052) and 5.5 g (0.032 mol) of 5-chloro-2,4-difluorobenzonitrile in 100 ml of dimethyl sulphoxide and the mixture is subsequently heated at 100°C for 36 hours. For working-up, the cooled reaction mixture is poured into water, the pH is brought to 2 using dilute hydrochloric acid, and the mixture is extracted repeatedly using dichloromethane. The combined organic phases are dried over sodium sulphate
20 and concentrated in vacuum. The residue is chromatographed over silica gel (eluent: dichloromethane).

1.8 g (18 % of theory) of 2-(2-chloro-4-cyano-5-fluoro-phenyl)-4-methyl-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazolin-3-one of melting point 105°C are obtained.

Use Examples:

- 5 In the use examples, the compound (A) below is used as comparison substance:



2-(4-cyano-2,5-difluoro-phenyl)-5-methyl-4-propargyl-2,4-dihydro-3H-1,2,4-triazol-3-one (disclosed in EP-A 370332).

Example A

- 10 Pre-emergence test

Solvent: 5 parts by weight of acetone

Emulsifier: 1 part by weight of alkylaryl polyglycol ether

- To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier
- 15 is added and the concentrate is diluted with water to the desired concentration.

Seeds of the test plants are sown in normal soil and, after 24 hours, watered with the preparation of the active compound. It is expedient to keep constant the amount of water per unit area. The concentration of the active compound in the preparation is of

no importance, only the amount of active compound applied per unit area being decisive. After three weeks, the degree of damage to the plants is rated in % damage in comparison to the development of the untreated control. The figures denote:

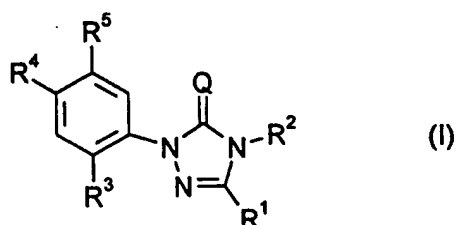
0% = no action (like untreated control)

5 100% = total destruction

In this test, clearly superior activity compared with the prior art in barley (0-10 %) and wheat (0 %) crops is shown, for example, by the compounds of Preparation Examples 4, 13, 17, 25, 26, 37 and 41 against weeds such as Digitaria (80-95 %), Abutilon (100 %), Chenopodium (95-100 %), Galinsoga (70-100 %), Matricaria
10 (60-100 %), Portulaca (70-100 %), Solanum (80-100 %) and Viola (80-100 %).

Patent Claims

1. New substituted triazolinones of the general formula (I)



in which

- 5 Q represents oxygen or sulphur,
- R¹ represents halogenoalkyl,
- R² represents hydrogen, amino, cyano, alkyl, alkenyl, alkynyl, halogenoalkyl, halogenoalkenyl, halogenoalkinyl, alkoxyalkyl, alkylideneimino or in each case optionally substituted cycloalkyl or cycloalkylalkyl,
- 10 R³ represents hydrogen or halogen,
- R⁴ represents cyano or nitro, and
- R⁵ represents isocyano, thiocyanato, sulpho, halogenosulphonyl, alkylamino-oxy, dialkylaminooxy, alkylideneaminooxy, cycloalkylideneaminooxy, or in each case optionally substituted cycloalkenyloxy or heterocyclyloxy, or
- 15 represents one of the following groups which are bonded via nitrogen or oxygen

-NR⁶R⁷, -N=CR⁸R⁹, -O-CO-R¹⁰, -O-CS-R¹⁰, -O-CHR¹¹-P(O)(OR¹²)₂, where

- 5 R^6 represents hydrogen, or represents alkyl, alkenyl or alkynyl, each of which is optionally substituted by halogen, or represents the group $-CO-R^{13}$ in which
- R^{13} represents hydrogen, or represents alkyl or alkoxy, each of which is optionally substituted by halogen or alkoxy, or represents amino, alkylamino or dialkylamino, or represents in each case optionally substituted cycloalkyl, cycloalkylalkyl, aryl, arylalkyl or heterocyclalkyl,
- 10 R^7 represents the group $-(CO)_n-R^{13}$ in which
- R^{13} has the abovementioned meaning and
- n represents the numbers 1 or 2,
- R^8 represents hydrogen, alkyl or alkoxy,
- R^9 represents alkoxy, alkylamino or dialkylamino,
- 15 R^{10} represents alkyl which is optionally substituted by halogen, or represents alkoxy, or represents alkylamino, or represents dialkylamino or represents optionally substituted aryl,
- R^{11} represents alkyl or cycloalkyl, each of which is optionally substituted by halogen, or represents in each case optionally substituted aryl or heterocycl, and
- 20 R^{12} represents alkyl.

2. New substituted triazolinones of the general formula (I) according to Claim 1, characterized in that

Q represents oxygen or sulphur,

5 R¹ represents straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, in particular fluorine, chlorine, bromine or iodine,

10 R² represents hydrogen, amino, cyano, straight-chain or branched alkyl having 1 to 8 carbon atoms, in each case straight-chain or branched alkenyl or alkynyl, each of which has 2 to 6 carbon atoms, straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, in particular fluorine, chlorine, bromine or iodine, or represents in each case straight-chain or branched halogenoalkenyl or halogenoalkynyl, each of which has 2 to 6 carbon atoms and 1 to 11 identical or different halogen atoms, in particular
15 fluorine, chlorine, bromine or iodine, or represents straight-chain or branched alkoxyalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, or represents straight-chain or branched alkylideneimino having 1 to 8 carbon atoms, or represents cycloalkyl or cycloalkylalkyl, each of which has 3 to 8 carbon atoms in the cycloalkyl moiety and, if appropriate, 1 to 4 carbon atoms in the straight-chain or
20 branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted in the cycloalkyl moiety by identical or different halogen substituents, in particular fluorine, chlorine, bromine and/or iodine,

25 R³ represents hydrogen, fluorine, chlorine, bromine or iodine,

R⁴ represents cyano or nitro, and

5 R^5 represents isocyano, thiocyanato, sulpho, halogenosulphonyl, C_1 - C_6 -alkylaminooxy, di(C_1 - C_4 -alkyl)-aminooxy, C_1 - C_6 -alkylideneaminooxy or C_5 - C_6 -cycloalkylideneaminooxy, or represents C_5 - C_6 -cycloalkenyloxy, perhydrofuranyloxy or perhydropyranyloxy, each of which is optionally substituted by halogen, C_1 - C_4 -alkyl or C_1 - C_4 -alkoxy, or represents one of the following groups which are bonded via nitrogen or oxygen

$-NR^6R^7$, $-N=CR^8R^9$, $-O-CO-R^{10}$, $-O-CS-R^{10}$, $-O-CHR^{11}-P(O)(OR^{12})_2$,
where

10 R^6 represents hydrogen, or represents alkyl, alkenyl or alkynyl, each of which has up to 6 carbon atoms and each of which is optionally substituted by fluorine and/or chlorine, or represents the group $-CO-R^{13}$ in which

15 R^{13} represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 6 carbon atoms and each of which is optionally substituted by fluorine, chlorine or C_1 - C_4 -alkoxy, or represents amino, or represents alkylamino or dialkylamino, each of which has 1 to 6 carbon atoms in the alkyl moieties, or represents in each case optionally substituted C_3 - C_6 -cycloalkyl, C_3 - C_6 -cycloalkyl- C_1 - C_4 -alkyl, phenyl, naphthyl, phenyl- C_1 - C_4 -alkyl, furyl, thienyl or pyridyl, suitable substituents in each case being:

20 halogen, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 4 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl, each of which

25

5 has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in each case straight-chain or branched alkoxycarbonyl or alkoxyiminoalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, and phenyl which is optionally
10 monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen and/or straight-chain or branched alkyl or alkoxy, each of which has 1 to 4 carbon atoms, and/or straight-chain or branched halogenoalkyl or halogenoalkoxy, each of which has 1 to 4 carbon atoms and
15 1 to 9 identical or different halogen atoms;

R^7 represents the group $-(CO)_n-R^{13}$ in which

R^{13} has the meaning given above as being preferred and

n represents the numbers 1 or 2,

15 R^8 represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 6 carbon atoms,

R^9 represents alkoxy, alkylamino or dialkylamino, each of which has 1 to 6 carbon atoms in the alkyl groups,

20 R^{10} represents alkyl having 1 to 6 carbon atoms which is optionally substituted by fluorine and/or chlorine, or represents alkoxy, alkylamino or dialkylamino or dialkylamino, each of which has 1 to 6 carbon atoms in the alkyl groups, or represents optionally substituted phenyl, preferred phenyl substituents being, again, those which have been mentioned above for R^6 as being preferred,

5 R¹¹ represents alkyl having 1 to 6 carbon atoms or cycloalkyl having 3 to 6 carbon atoms, in each case optionally substituted by fluorine and/or chlorine, or represents in each case optionally substituted phenyl, naphthyl, furyl, thienyl or pyridyl, preferred phenyl substituents being, again, those which have been mentioned above for R⁶ as being preferred, and

R¹² represents alkyl having 1 to 6 carbon atoms.

3. New substituted triazolinones of the general formula (I) according to Claim 1, characterized in that

Q represents oxygen or sulphur,

10 R¹ represents straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in particular fluorine, chlorine, bromine or iodine,

15 R² represents hydrogen, amino, cyano, straight-chain or branched alkyl having 1 to 4 carbon atoms, in each case straight-chain or branched alkenyl or alkynyl, each of which has 3 to 4 carbon atoms, straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in particular fluorine or chlorine, in each case straight-chain or branched halogenoalkenyl or halogenoalkynyl, each of which has 3 to 4 carbon atoms and 1 to 5 identical or different halogen atoms, in particular fluorine or chlorine, or represents straight-chain or branched alkoxyalkyl having in each case 1 to 3 carbon atoms in the individual alkyl moieties, or represents straight-chain or branched alkylideneimino having 1 to 4 carbon atoms, or represents cycloalkyl or cycloalkylalkyl, each of which has 3 to 6 carbon atoms in the cycloalkyl moiety and, if appropriate, 20 1 to 3 carbon atoms in the straight-chain or branched alkyl moiety and each 25

of which is optionally monosubstituted or polysubstituted in the cycloalkyl moiety by identical or different halogen substituents, in particular fluorine or chlorine,

R³ represents hydrogen, fluorine or chlorine,

5 R⁴ represents cyano or nitro, and

R⁵ represents isocyano, thiocyanato, sulpho, chlorosulphonyl, C₁-C₄-alkyl-aminooxy, di-(C₁-C₃-alkyl)-aminooxy, C₁-C₄-alkylidene-aminooxy, C₅-C₆-cycloalkylideneaminooxy, C₅-C₆-cycloalkenyloxy, tetrahydrofuranyl-oxy, perhydropyranyloxy or one of the following groups which are bonded
10 via nitrogen or oxygen

-NR⁶R⁷, -N=CR⁸R⁹, -O-CO-R¹⁰, -O-CS-R¹⁰, -O-CHR¹¹-P(O)(OR¹²)₂ where

R⁶ represents hydrogen, or represents alkyl having 1 to 4 carbon atoms which is optionally substituted by fluorine, or represents the group -CO-R¹³ in which

15 R¹³ represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 4 carbon atoms and each of which is optionally substituted by fluorine, chlorine, methoxy or ethoxy, or represents amino, or represents alkylamino or dialkylamino, each of which has 1 to 4 carbon atoms in the alkyl moieties, or represents in
20 each case optionally substituted cyclopropyl, cyclopentyl cyclohexyl, cyclopropylmethyl, cyclopentylmethyl, cyclohexylmethyl, phenyl, benzyl, phenylethyl, furyl, thienyl or pyridyl, suitable substituents in each case being:
fluorine, chlorine, bromine, cyano, nitro, in each case straight-

chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or
 alkylsulphonyl, each of which has 1 to 4 carbon atoms, in each
 case straight-chain or branched halogenoalkyl, halogenoalkoxy,
 halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkyl-
 5 sulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9
 identical or different halogen atoms, in particular fluorine and/or
 chlorine atoms, in each case straight-chain or branched alkoxy-
 carbonyl or alkoxyiminoalkyl, each of which has 1 to 4 carbon
 atoms in the individual alkyl moieties, and phenyl which is
 10 optionally monosubstituted or polysubstituted by identical or
 different substituents from the series consisting of fluorine,
 chlorine, bromine and/or straight-chain or branched alkyl or
 alkoxy, each of which has 1 to 4 carbon atoms, and/or straight-
 chain or branched halogenoalkyl or halogenoalkoxy, each of
 15 which has 1 to 4 carbon atoms and 1 to 9 identical or different
 halogen atoms, in particular fluorine and/or chlorine atoms;

R^7 represents the group $-(CO)_n-R^{13}$ in which

R^{13} has the meaning given above as being particularly preferred and

n represents the numbers 1 or 2,

20 R^8 represents hydrogen, or represents alkyl or alkoxy, each of which has 1 to 4 carbon atoms,

R^9 represents alkoxy, alkylamino or dialkylamino, each of which has 1 to 4 carbon atoms in the alkyl groups,

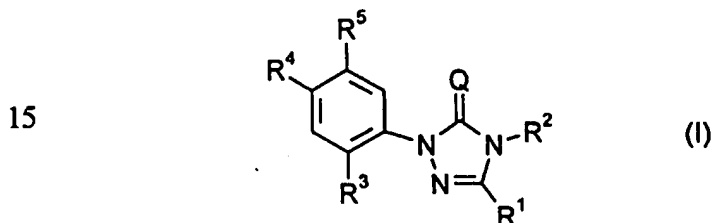
R^{10} represents alkyl having 1 to 4 carbon atoms which is optionally substituted

5 by fluorine and/or chlorine, or represents alkoxy, alkylamino or dialkylamino, each of which has 1 to 4 carbon atoms in the alkyl groups, or represents optionally substituted phenyl, preferred phenyl substituents being, again, those which have been mentioned above for R⁶ as being particularly preferred,

10 R¹¹ represents alkyl having 1 to 4 carbon atoms or cycloalkyl having 3 to 6 carbon atoms, in each case optionally substituted by fluorine and/or chlorine, or represents in each case optionally substituted phenyl, naphthyl, furyl, thienyl or pyridyl, preferred phenyl substituents being, again, those which have been mentioned above for R⁶ as being particularly preferred, and

R¹² represents alkyl having 1 to 4 carbon atoms.

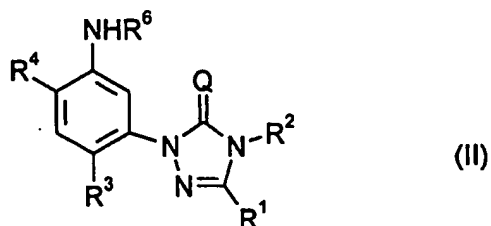
4. Process for the preparation of new substituted triazolinones of the general formula (I)



in which

R¹, R², R³, R⁴, R⁵ and Q have the meanings given in Claim 1, characterized in that

(a) aminoaryltriazolinones of the general formula (II)



in which

Q, R¹, R², R³, R⁴ and R⁶ have the abovementioned meanings

are reacted with acid halides of the general formula (III)



in which

n and R¹³ have the abovementioned meaning and

X¹ represents halogen,

or with ortho esters of the general formula (IV)



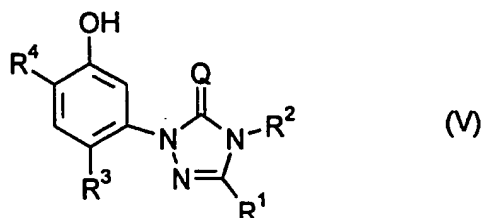
in which

R⁸ and R⁹ have the abovementioned meanings,

if appropriate in the presence of a reaction auxiliary and if appropriate in the presence of a diluent,

or in that

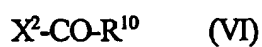
(b) hydroxyaryltriazolinones of the general formula (V)



in which

5 Q, R¹, R², R³ and R⁴ have the abovementioned meanings

are reacted with acid halides of the general formula (VI)



in which

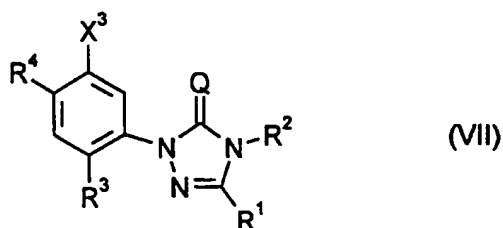
R¹⁰ has the abovementioned meaning and

10 X² represents halogen,

if appropriate in the presence of an acid acceptor and if appropriate in the presence of a diluent,

or in that

(c) halogenoaryltriazolinones of the general formula (VII)



in which

Q, R¹, R², R³ and R⁴ have the abovementioned meanings and

X³ represents halogen,

5 are reacted with hydroxyalkylphosphonic esters of the general formula (VIII)



in which

R¹¹ and R¹² have the abovementioned meanings,

10 if appropriate in the presence of an acid acceptor and if appropriate in the presence of a diluent.

5. Herbicidal compositions, characterized in that they comprise at least one substituted triazolinone of the general formula (I) according to Claims 1 to 5.
6. Method of combating undesirable plants, characterized in that substituted triazolinones of the general formula (I) according to Claims 1 to 5 are allowed to act on plants and/or their environment.
- 15 7. Use of substituted triazolinones of the general formula (I) according to Claims 1

to 5 for combating undesirable plants.

8. Process for the preparation of herbicidal compositions, characterized in that substituted triazolinones of the general formula (I) according to Claims 1 to 5 are mixed with extenders and/or surface-active substances

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